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The use of Magnetic Sector Secondary Ion Mass Spectrometry to investigate ^{14}C distribution in Magnox reactor core graphite.

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Abstract

Large quantities of irradiated graphite containing carbon-14 will arise from the decommissioning of the UK's Magnox power stations. Magnetic sector secondary ion mass spectrometry (MS-SIMS) has been used to investigate the distribution of this radioisotope within trepanned graphite samples from a Magnox nuclear power station. This showed that a carbonaceous deposit found on channel wall samples had an apparent enrichment of ^{14}C compared to underlying graphite. Samples without this deposit gave ^{14}C concentrations below the limits of detection of the instrument. The methodology used ensured that possible interferences between ^{14}C species and oxygen-bearing species were eliminated from the analysis.

Background

The decommissioning of the first generation of gas-cooled, graphite-moderated reactors in the United Kingdom will lead to approximately 45,000 m^3 of graphite waste requiring disposal [1]. The majority of this will be classified as intermediate level waste (ILW) [2] and contains the long lived radionuclide ^{14}C, which is important in safety assessments of a geological disposal facility (GDF) in the UK [3]. This work investigates the use of MS-SIMS to determine the distribution of this radioisotope in irradiated graphite from a Magnox reactor. A carbonaceous deposit formed during plant operation [4] on the fuel channel walls has been investigated to establish if there is any relative enrichment in ^{14}C compared to the underlying graphite. The ^{14}C found in such a deposit could be more easily released after disposal than ^{14}C within the bulk material.

Results

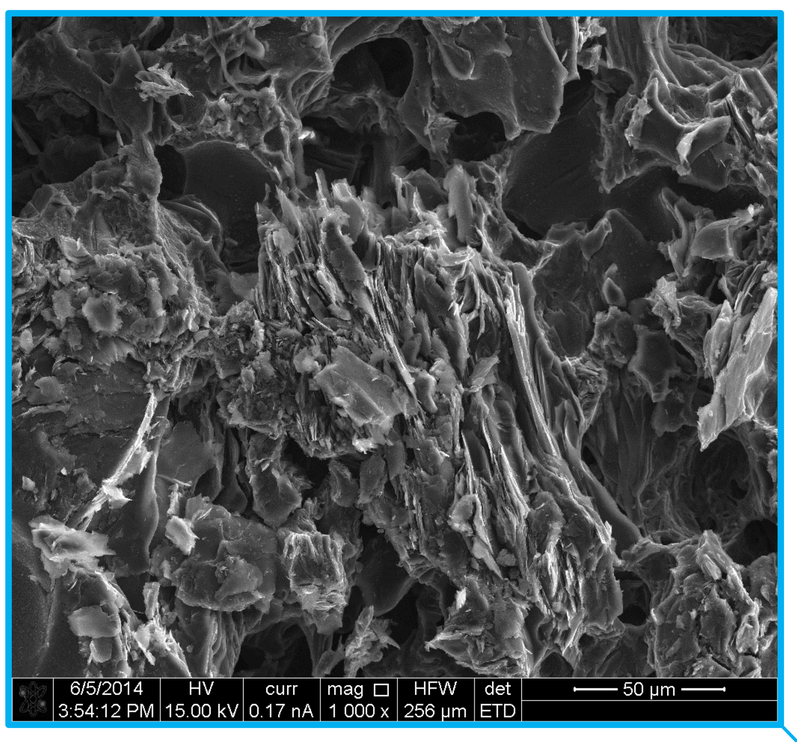

Figure 2(b):Electron micrograph showing the structure of inner brick (non-channel wall face) of irradiated graphite.

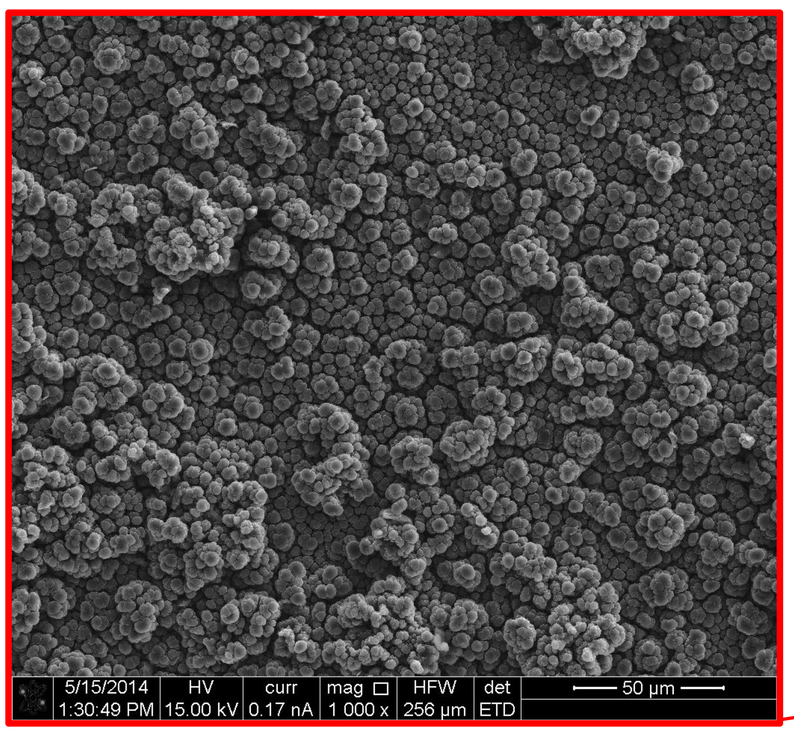

Figure 2(c):Electron micrograph showing the carbonaceous deposit found on a channel wall face of irradiated graphite.

Experimental

Samples

Irradiated pile grade A (PGA) graphite samples trepanned from Oldbury power station were examined using SEM and MS-SIMS. The cylindrical trepan had previously been cut into 3 slices by National Nuclear Laboratory (NNL). No further preparation was performed before analysis. Virgin (PGA) graphite material which was surplus from the commissioning of Wylfa power station was provided by Magnox Ltd. This material was machined using a diamond cutting wheel to similar dimensions as the trepanned irradiated samples.

Instrument

An in-house built MS-SIMS comprising of a gallium ion gun and a double focusing electrostatic/magnetic sector mass analyser was used for the analysis. The instrument was used in negative mode with a gallium ion beam of 3nA current and 25 keV energy throughout. A sample bias voltage of 4kV was used during analysis. The area of analysis was approximately 60µm x 60µm. A Helios NanoLab 600i DualBeam integrated system (FEI, Oregon USA) was used to obtain secondary electron micrographs of sample surfaces.

Methodology

Depth profiles (t=1800s) were obtained from five different areas of each trepan sample face (six faces per sample set). Due to the isobaric interference from CH_2^- at m/z 14 the C_2^- signal at m/z 28 was used. The average ratio between m/z 28 and m/z 24 as well as m/z 16 and m/z 24 were calculated between 200-1800 seconds, Figure 2(a). As the signal at m/z 24 ($^{12}C_2$) was believed to be steady it was used as a baseline. Investigation into possible CO^- interference at m/z 28 was performed by collecting identical depth profiles and calculations on virgin PGA graphite that had been incrementally exposed to experimental grade oxygen from a total chamber pressure of $2x10^{-8}$mbar up to a maximum chamber pressure of 1 x 10^{-6}mbar in order to generate CO^- species on the sample surface. Figure 1 shows the signal from mass 28 from CO^- as a function of the oxygen signal (both normalised to the mass 24 signal from C_2^-, assumed to be constant). The ^{14}C signal from the irradiated samples (Figure 2(a)) was inferred by observing the deviation of the 28/24 ratio from the line. Micrographs highlight the difference in appearance between the underlying, radiolytically oxidised graphite and the carbonaceous coating found on channel wall surfaces, Figures 2(b) and 2(c) respectively.

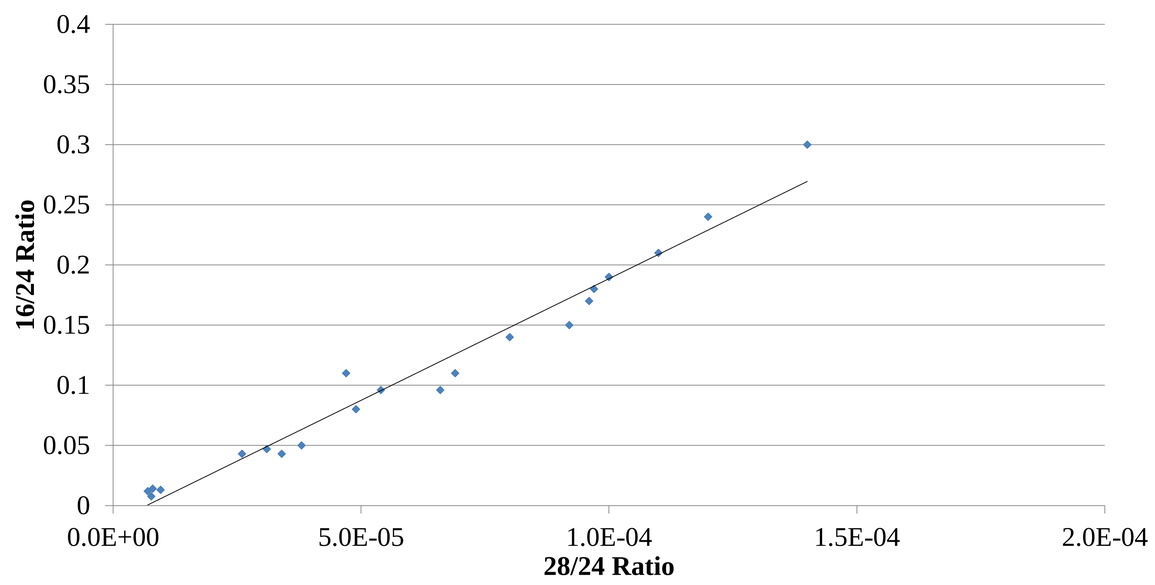

Figure 1:The relationship between the 28:24 and 16:24 ratio for virgin PGA exposed incrementally to oxygen during analysis. The m/z 28 signal is assumed to be from CO^- species generated on the surface.

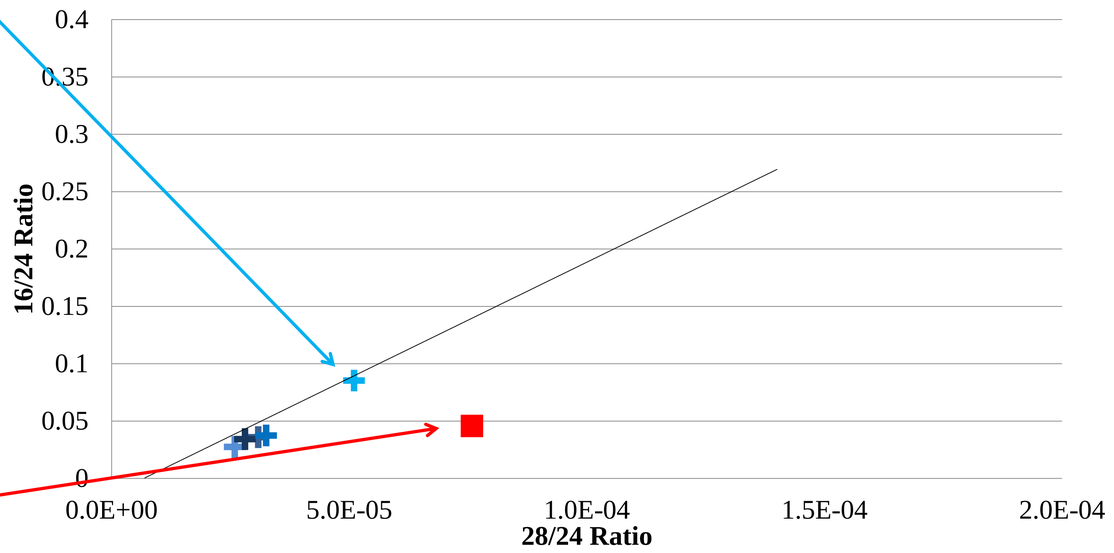

Figure 2(a): Comparison of 28:24 to 16:24 ratios for a set of trepanned graphite samples (Red square shows channel wall face and blue crosses show inner brick samples). Solid line is from Figure 1.

Conclusions

- The MS-SIMS technique is of use in investigating the distribution of ^{14}C in irradiated graphite.
- Although the presence of oxygen contributes to the signal at m/z 28, a comparison of the m/z 28:24 ratio to the m/z 16:24 ratio allows the ^{14}C contribution to be distinguished.
- An apparent enrichment of ^{14}C in the carbonaceous deposits found on channel wall face samples from an Oldbury Magnox reactor is observed.
- The concentration of ^{14}C within the bulk graphite (inner brick) is below the limit of detection for this technique

Future work

This work will continue to use this technique to examine further trepanned samples from various locations within the reactor core to investigate any variation in the apparent 14C enrichment of carbonaceous deposit with position

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